5720

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☐ 1. Document ID: US 5254788 A

L9: Entry 1 of 2 File: USPT Oct 19, 1993

US-PAT-NO: 5254788
DOCUMENT-IDENTIFIER: US 5254788 A
** See image for Certificate of Correction **

TITLE: Process for the production of olefins from light paraffins

DATE-ISSUED: October 19, 1993

INVENTOR-INFORMATION:

NAME	CITY	STATE	ZIP CODE	COUNTRY
Gartside; Robert J.	Summit	NJ		
Johnson; Axel R.	N. Babylon	NY		

US-CL-CURRENT: 585/659; 585/654, 585/660, 585/661

Full | Title | Citation | Front | Review | Classification | Date | Reference | Sequences | Attachments | Claims | KWIC | Draw. De

☐ 2. Document ID: US 5220093 A

L9: Entry 2 of 2 File: USPT Jun 15, 1993

US-PAT-NO: 5220093
DOCUMENT-IDENTIFIER: US 5220093 A
** See image for Certificate of Correction **

TITLE: Process for production of olefins from mixtures of light paraffins

DATE-ISSUED: June 15, 1993

INVENTOR-INFORMATION:

NAME	CITY	STATE	ZIP CODE	COUNTRY
Gartside; Robert J.	Summit	NJ		
Johnson; Axel R.	North Babylon	NY		

US-CL-CURRENT: 585/661; 585/654

Full | Title | Citation | Front | Review | Classification | Date | Reference | Sequences | Attachments | Claims | KWIC | Draw. De

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Terms	Documents
L8 and gas separation	2

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L12: Entry 8 of 11 File: USPT Sep 12, 2000

US-PAT-NO: 6117328
DOCUMENT-IDENTIFIER: US 6117328 A

TITLE: Adsorbent-filled membranes for pervaporation

DATE-ISSUED: September 12, 2000

INVENTOR-INFORMATION:

NAME	CITY	STATE	ZIP CODE	COUNTRY
Sikdar; Subhas K.	Cincinnati	OH		
Ji; Wenchang	Natick	MA		
Wang; Sun-tak	Cincinnati	OH		

US-CL-CURRENT: 210/640; 210/500.27, 210/502.1, 210/653, 95/45, 95/50, 96/4

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EAST Search History

Ref #	Hits	Search Query	DBs	Default Operator	Plurals	Time Stamp
L1	4496	zeolite membrane	USPAT; EPO; DERWENT	AND	ON	2006/05/17 14:47
L2	295	1 and asymmetric	USPAT; EPO; DERWENT	AND	ON	2006/05/17 14:48
L3	295	2 asymmetric membrane	USPAT; EPO; DERWENT	AND	ON	2006/05/17 14:48
L4	253	3 and carbon	USPAT; EPO; DERWENT	AND	ON	2006/05/17 14:49
L5	0	4 and coating and carboniztion	USPAT; EPO; DERWENT	AND	ON	2006/05/17 14:49
L6	12	4 and carbonization	USPAT; EPO; DERWENT	AND	ON	2006/05/17 14:49

US-PAT-NO: 6921428

DOCUMENT-IDENTIFIER: US 6921428 B2

TITLE: Device and method for separating and collecting halide gas

DATE-ISSUED: July 26, 2005

INVENTOR-INFORMATION:

NAME	CITY	STATE	ZIP CODE	COUNTRY
Yamamoto; Osamu	Kyoto	N/A	N/A	JP
Kinouchi; Masayuki	Ube	N/A	N/A	JP
Takada; Naoki	Ube	N/A	N/A	JP
Kase; Yoji	Ichihara	N/A	N/A	JP
Kikuchi; Masao	Ichihara	N/A	N/A	JP

ASSIGNEE INFORMATION:

NAME	CITY	STATE	ZIP CODE	COUNTRY	TYPE CODE
Ube Industries, Ltd.	Yamaguchi	N/A	N/A	JP	03

APPL-NO: 10/466188

DATE FILED: July 14, 2003

FOREIGN-APPL-PRIORITY-DATA:

COUNTRY	APPL-NO	APPL-DATE
JP	2001-016571	January 25, 2001
JP	2001-393642	December 26, 2001
JP	2002-010262	January 18, 2002

PCT-DATA:
APPL-NO: PCT/JP02/00470
DATE-FILED: January 23, 2002
PUB-NO: WO02/05882
PUB-DATE: Aug 1, 2002
371-DATE: Jul 14, 2003
102(E)-DATE: July 14, 2003

INT-CL-ISSUED: [07] B01D053/22 , B01D069/08 , B01D071/64

INT-CL-CURRENT:

TYPE	IPC	DATE
CIPS	B01D53/22	20060101
CIPS	B01D71/02	20060101
CIPS	B01D71/00	20060101
CIPS	B01D61/58	20060101
CIPS	B01D71/64	20060101

US-CL-ISSUED: 95/45 , 95/47 , 96/8 , 96/9 , 96/10 , 96/14

US-CL-CURRENT: 95/45, 95/47 , 96/10 , 96/14 , 96/8 , 96/9

FIELD-OF-CLASSIFICATION-SEARCH: 95/45; 95/47 ; 96/4 ; 96/7-10 ; 96/14
See application file for complete search history

REF-CITED:

U.S. PATENT DOCUMENTS

PAT-NO	ISSUE-DATE	PATENTEE-NAME	US-CL		
4264338	April 1981	Null	95/47	N/A	N/A
5064447	November 1991	Lee	N/A	N/A	N/A
5281253	January 1994	Thompson	95/22	N/A	N/A
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N/A

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N/A				
5976222	November 1999	Yang et al.	95/45	N/A
N/A				
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N/A				
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N/A				
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N/A				
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N/A				
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N/A				
2003/0047068	March 2003	Brazier	95/45	N/A
N/A				

FOREIGN PATENT DOCUMENTS

FOREIGN-PAT-NO	PUBN-DATE	COUNTRY	US-CL
0 754 487	January 1997	EP	
1 034 836	September 2000	EP	
1 080 774	March 2001	EP	
A 10-298118	November 1998	JP	
A 11-345545	December 1999	JP	
A 2000-140558	May 2000	JP	
2000-140576	May 2000	JP	
A 2000-185212	July 2000	JP	
WO 00/71232	November 2000	WO	

OTHER PUBLICATIONS

H. Hama et al., "Application Problems of SF.sub.6 /N.sub.2 Mixtures to Gas Insulated Bus," Gaseous Dielectrics VIII, Ed. by Christophorou and Olthoff, 1998, pp. 353-359.

ART-UNIT: 1724

PRIMARY-EXAMINER: Spitzer; Robert H.

ATTY-AGENT-FIRM: Young & Thompson

ABSTRACT:

A halide gas separating and collecting device for separating and collecting halide gas from mixed gas containing the halide gas, wherein at least first and second stage separating **membrane** modules are stacked in multiple stages. The method comprises: feeding the mixed gas to the inlet of the first stage separating **membrane** module, feeding the gas passed through the previous separating **membrane** module to the inlets of the second and subsequent stage separating **membrane** modules, recycling the gas unpassed through the second and subsequent stage separating **membrane** modules to the inlet of the first stage separating **membrane** module, and controlling the flow of gas unpassed through the first stage separating **membrane** module by a control valve connected to the unpassed gas outlet of the first stage separating **membrane** module, whereby the halide gas can be separated and collected as unpassed gas at a high density and a high collection rate.

17 Claims, 2 Drawing figures

Exemplary Claim Number: 1

Number of Drawing Sheets: 2

---------- KWIC ---------

Abstract Text - ABTX (1):

A halide gas separating and collecting device for separating and collecting halide gas from mixed gas containing the halide gas, wherein at least first and second stage separating **membrane** modules are stacked in multiple stages. The method comprises: feeding the mixed gas to the inlet of the first stage separating **membrane** module, feeding the gas passed through the previous separating **membrane** module to the inlets of the second and subsequent stage separating **membrane** modules, recycling the gas unpassed through the second and subsequent stage separating **membrane** modules to the inlet of the first stage separating **membrane** module, and controlling the flow of gas unpassed through the first stage separating **membrane** module by a control valve connected to the unpassed gas outlet of the first stage separating **membrane** module, whereby the halide gas can be separated and collected as unpassed gas at a high density and

a high collection rate.

Brief Summary Text - BSTX (5):

Nevertheless because satisfactory substitutes have not been discovered as yet, there still remain some applications in which halide gases continue being used. For example, there is found no satisfactory substitute for electrically insulating gas for gas-insulated electrical equipment, such as gas-insulated switchgears, gas circuit breakers, and gas-insulated transformers. Hence in an attempt to reduce use of halide gases, electric power industries have developed replacement of pure halide gases for electric insulation with mixed gases comprising a halide gas, e.g., SF.sub.6 (sulfur hexafluoride), Freon or **carbon** tetrachloride, and a diluting gas such as N.sub.2 gas. SF.sub.6 gas is characterized by high dielectric strength upon being pressurized and by lower liquefying temperature than Freon or **carbon** tetrachloride gas, which enables use in low temperatures. Therefore, a mixed gas comprising SF.sub.6 gas and a diluting gas is a highly promising substitute as an electrically insulating gas.

Brief Summary Text - BSTX (14):

JP-A-11-345545 proposes using a gas separation **membrane** selected from a polyimide **membrane, a carbon membrane, and a zeolite membrane** as an effective substitute for the pressure cooling method in the separation and recovery of SF.sub.6 gas. According to this method SF.sub.6 gas is separated and recovered as non-permeate gas of the gas separation **membrane**. It is true that SF.sub.6 gas has a smaller rate of permeation than other gas of a mixed gas (e.g., N.sub.2 gas), but this does not mean that SF.sub.6 gas is incapable of passing through a gas separation **membrane** at all. SF.sub.6 gas, while not much compared with other gas (e.g., N.sub.2 gas), also enters a permeate gas stream through a gas separation **membrane** together with the other gas. The SF.sub.6 in the permeate gas results in recovery loss. This recovery loss increases with an attempt to increase the purity of SF.sub.6 gas in the non-permeate. Therefore, it is difficult to separate and recover SF.sub.6 gas with a high concentration (high purity) at a high recovery rate.

Brief Summary Text - BSTX (15):

JP-A-2000-140558 discloses a method and an apparatus for separating and recovering SF.sub.6 gas from an SF.sub.6 -containing mixed gas by use of an aromatic polyimide separation **membrane**. The publication is silent to a specific process or a specific apparatus with which a halide gas can be recovered with a reduced recovery loss and with a high purity enough to be reused.

Brief Summary Text - BSTX (16):
JP-A-2000-185212 discloses a method and an apparatus for separating and recovering a perfluorocompound gas from a perfluorocompound-containing mixed gas by use of an **asymmetric carbon membrane** obtained by carbonizing an **asymmetric** polyimide film. This publication has no mention of a specific process or a specific apparatus with which a halide gas can be recovered with a reduced recovery loss and with a high purity enough to be reused.

Brief Summary Text - BSTX (17):
JP-A-10-128034 discloses a method of separating and recovering a fluorochemical gas from a mixed gas containing a diluting gas and the fluorochemical gas by use of gas separation **membranes,** wherein the non-permeate gas of a first gas separation **membrane** is led to a second gas separation **membrane,** and the non-permeate gas of the second gas separation **membrane** is collected as a separated and recovered fluorochemical gas.

Brief Summary Text - BSTX (18):
JP-A-9-103633 proposes a method and an apparatus for separating and recovering a perfluorocompound gas from a gas mixture using a gas separation **membrane** made of a glassy polymer, wherein the non-permeate gas of a first gas separation **membrane** is led to a second gas separation **membrane,** and the non-permeate gas of the second gas separation **membrane** is collected as a separated and recovered perfluorocompound gas.

Brief Summary Text - BSTX (19):
JP-A-10-298118 discloses a method and a system for separating and recovering a perfluorocompound gas from a gas mixture using gas separation **membranes** including a **carbon** sieve **membrane,** wherein the non-permeate stream of a first gas separation **membrane** is sent to a second gas separation **membrane,** and the non-permeate stream of the second gas separation **membrane** is collected as a separated and recovered perfluorocompound gas.

Brief Summary Text - BSTX (20):
These methods using a plurality of gas separation **membranes** are suitable to recover a fluorochemical gas or a perfluorocompound gas with a higher purity from a mixed gas. However, the methods also involve the recovery loss problem due to passage of a fluorochemical gas or a perfluorocompound gas through the first gas separation **membrane** together with a diluting gas. Electrically insulating gas comprising a mixture of, for instance, SF.sub.6 gas and a diluting gas usually has a relatively high SF.sub.6 content ranging 3 to 60% by volume. When these methods are applied to the separation and recovery of SF.sub.6 gas from such an electrically insulating gas, it is difficult to

separate and recover SF.sub.6 gas in high concentration (high purity) at high recovery rate since an unignorable amount of SF.sub.6 gas is emitted together with the permeate gas of the first gas separation **membrane**.

Brief Summary Text - BSTX (21):

The technique using a gas separation **membrane** is superior to the pressure cooling method for the separation and recovery of a halide gas from a halide gas-containing mixed gas. In the light of the above-mentioned problem notwithstanding the superiority, it has been desired to develop an improved apparatus and an improved method capable of reducing the recovery loss of a halide gas and increasing the purity of the recovered gas to a level enough to be reused.

Brief Summary Text - BSTX (24):

The present invention relates to an apparatus for separating and recovering a halide gas comprising at least first and second stage separation **membrane** modules which is characterized in that the first stage separation **membrane** module has a gas inlet thereof connected to a source of a mixed gas containing a halide gas and a non-permeate gas outlet thereof connected via a gas flow control valve to a gas recovery container, and the second stage separation **membrane** module has a gas inlet thereof connected to the permeate gas outlet of the first separation **membrane** module and a non-permeate gas outlet thereof connected to the source of the halide gas-containing mixed gas.

Brief Summary Text - BSTX (25):

The apparatus of the present invention includes the following preferred embodiments. 1) The non-permeate gas outlet of the first stage separation **membrane** module is connected to the gas recovery container via the gas flow control valve and a gas concentration detector. 2) The gas concentration detector and the gas flow control valve are coupled by a controller into a configuration such that the gas flow control valve is controlled based on a measured value of the gas concentration detector so as to adjust the amount of the gas collected from the non-permeate gas outlet. 3) The separation **membrane** module is configured to be maintained at a constant temperature in a range of from 40 to 200.degree. C. by a heating unit. 4) The apparatus has at least three separation **membrane** modules, wherein each of the third and the following stage separation **membrane** modules has the gas inlet thereof connected to the permeate gas outlet of the immediately preceding separation **membrane** module and the non-permeate gas outlet thereof connected to the source of the halide gas-containing mixed gas. 5) The separation **membrane** module comprises an aromatic polyimide hollow fiber separation **membrane**. 6) The separation **membrane** module comprises a carbonized separation **membrane** obtained by

carbonizing a polymer. 7) The carbonized separation **membrane** is a partially carbonized hollow fiber separation **membrane** obtained by partially carbonizing an aromatic polyimide **asymmetric** hollow fiber **membrane**. 8) The mixed gas of the mixed gas source is an electrically insulating gas containing a halide gas and a diluting gas. 9) The halide gas is SF.sub.6 gas. 10) The mixed gas of the mixed gas source has a halide gas concentration of 3 to 60% by volume.

Brief Summary Text - BSTX (26):

The present invention also relates to a method of separating and recovering a halide gas from a mixed gas containing the halide gas through multiple stages using at least two separation **membrane** modules, which comprises the steps of feeding a mixed gas containing a halide gas to the gas inlet of the first stage separation **membrane** module, feeding, to the gas inlet of each of the second and the following stage separation **membrane** modules, a permeate gas of an immediately preceding stage separation **membrane** module, recycling the non-permeate gas of each of the second and the following stage separation **membrane** modules to the gas inlet of the first stage separation **membrane** module, and collecting the non-permeate gas of the first stage separation **membrane** module with a prescribed halide gas concentration while controlling the non-permeate gas flow from the first stage separation **membrane** module with a gas flow control valve connected to the non-permeate gas outlet of the first stage separation **membrane** module.

Brief Summary Text - BSTX (27):

The method of the present invention includes the following preferred embodiments. 11) A gas concentration detector and the gas flow control valve are connected to the non-permeate gas outlet of the first stage separation **membrane** module in such a configuration that the gas concentration detector measures the halide gas concentration of the non-permeate gas discharged from the non-permeate gas outlet of the first stage separation **membrane** module and that the gas flow control valve controls the non-permeate gas flow based on the measured value of the gas concentration detector thereby to collect the halide gas in a prescribed concentration. 12) The separation **membrane** module is configured to be maintained at a constant temperature in a range of from 40 to 200.degree. C. by a heating unit. 13) The separation **membrane** module comprises an aromatic polyimide hollow fiber separation **membrane**. 14) The separation **membrane** module comprises a carbonized separation **membrane** obtained by carbonizing a polymer. 15) The carbonized separation **membrane** is a partially carbonized hollow fiber separation **membrane** obtained by partially carbonizing an aromatic polyimide **asymmetric** hollow fiber **membrane**.

Detailed Description Text - DETX (8):

The separation **membrane** modules used in the present invention each comprise a container which has a gas inlet, a permeate gas outlet, and a non-permeate gas outlet and is equipped with a selectively gas permeable separation **membrane** to form a gas feed side space and a permeate side space isolated from each other. While the gas separation **membrane** may be a flat sheet **membrane,** a thin and small-diametered hollow fiber **membrane** is advantageous for size reduction of the apparatus and a larger **membrane** area that assures high separation efficiency and good economy. The gas separation **membrane** may be either homogeneous or heterogeneous as in a composite **membrane** or an asymmetrical **membrane** or may be either microporous or nonporous. Suitable hollow fiber **membranes** have a thickness of 10 to 500 .mu.M and an outer diameter of 50 to 2000 .mu.m.

Detailed Description Text - DETX (9):

Suitable examples of the separation **membrane** modules used in the present invention include those using gas separation **membranes** made of polymers, such as polyimide, polyether imide, polyamide, polyamide-imide, polysulfone, polycarbonate, and cellulosic polymers, and ceramics, such as **zeolite**. In particular, aromatic polyimide hollow fiber separation **membranes,** especially aromatic polyimide **asymmetric** hollow fiber separation **membranes** exhibit high separation performance between halide gases and diluting gases, e.g., N.sub.2 gas, high durability against halide gases, and sufficient heat resistance. Accordingly, an aromatic polyimide **asymmetric** hollow fiber separation **membrane,** when used in the first stage separation **membrane** module, achieves separation and recovery of a halide gas in a prescribed high concentration (high purity) as a non-permeate gas. Such a separation **membrane** allows elevation of the separation temperature to raise the separation efficiency. In addition, with this **membrane,** the separation operation can be continued for an extended period of time in a stable manner because the **membrane** is hardly susceptible to deterioration by halides or high temperature. For these reasons an aromatic polyimide hollow fiber separation **membrane,** especially an aromatic polyimide **asymmetric** hollow fiber separation **membrane** is particularly preferred in the present invention.

Detailed Description Text - DETX (10):

The aromatic polyimide separation **membrane** is preferably, but not necessarily, prepared by, for example, the processes taught in JP-A-5-68859 and JP-A-6-254367.

Detailed Description Text - DETX (11):

Separation **membrane** modules using a carbonized separation **membrane** obtained by carbonizing a polymer are also preferred. The carbonized separation

membranes obtained by polymer **carbonization** include a **carbon membrane** obtained by pyrolyzing a **membrane** formed of cellulose, etc. in high temperatures of 800 to 950.degree. C. for a long time as taught in U.S. Pat. No. 4,685,940; one consisting essentially of **carbon** that is obtained by subjecting the above-described **carbon membrane** to CVD processing, plasma treatment or oxidation treatment; and a porous hollow **carbon** fiber **membrane** consisting essentially of **carbon** that is obtained by spinning a mixture of polyacrylonitrile, a component decomposing at low temperatures of 600.degree. C. or lower, and a solvent into hollow fibers and treating the resulting hollow fibers at preferably 600 to 1200.degree. C. to cause the decomposable component to decompose and dissipate as disclosed in JP-A-2-74615.

Detailed Description Text - DETX (12):
The hollow fiber separation **membrane** to be used may be one having an **asymmetric** structure that is obtained by partially carbonizing an aromatic polyimide **asymmetric** hollow fiber **membrane** by low-temperature and/or short-time heat treatment as disclosed in JP-A-4-11933, JP-A-4-193334, JP-A-5-220360, and JP-A-2000-342944.

Detailed Description Text - DETX (13):
Carbonized separation **membranes** obtained by polymer **carbonization** exhibit high separation performance between halide gases and diluting gases, e.g., N.sub.2 gas, high durability against halide gases, and sufficient heat resistance. Accordingly, the carbonized separation **membrane** obtained by polymer **carbonization,** when used in the first stage separation **membrane** module, achieves separation and recovery of a halide gas in a prescribed high concentration (high purity) as a non-permeate gas. Such a separation **membrane** allows elevation of the separation temperature to raise the separation efficiency. In addition, with this **membrane,** the separation operation can be continued for an extended period of time in a stable manner because the **membrane** is hardly susceptible to deterioration by halides or high temperature.

Detailed Description Text - DETX (14):
Inter alia, hollow fiber separation **membranes with an asymmetric** structure which are obtained by partial **carbonization** of aromatic polyimide **asymmetric** hollow fiber **membranes** are highly advantageous in view of ease of fabrication and economy. That is, the aromatic polyimide **asymmetric** hollow fiber **membranes** as precursors are easy to fabricate, and the processing step involved is partial **carbonization** that can be accomplished by heating at a relatively low temperature for a short period of time. Besides, the hollow fiber separation **membranes** of this type exhibit extremely high separation performance and are easily designed to have relatively small diameters while retaining mechanical

strength required for industrial modularization. It is easy with this type of hollow fiber separation **membranes** to provide separation **membrane** modules having high separation performance and a large effective **membrane** area, with which a halide gas can be separated and recovered with extremely high efficiency.

Detailed Description Text - DETX (15):

For these reasons a separation **membrane** module having a hollow fiber separation **membrane** obtained by partially carbonizing an aromatic polyimide **asymmetric** hollow fiber separation **membrane** is particularly suitable for use in the present invention.

Detailed Description Text - DETX (16):

Where the separation **membrane** module used in the present invention is composed of the hollow fiber **membrane,** the module generally has the following structure. A large number (e.g., several hundreds to several hundreds of thousands) of hollow fiber **membranes** are gathered into a bundle. The hollow fibers of the bundle are firmly fixed to each other at one or both end portions thereof with a curing resin (e.g., an epoxy resin), a thermoplastic resin (e.g., a polyamide resin), etc. to make a hollow fiber separation **membrane** element with both ends of the individual fibers open. One or more than one elements thus prepared are set in a container having at least a gas inlet, a permeate gas outlet, and a non-permeate gas outlet in such a manner that the space leading to the inside of the individual hollow fibers and the space leading to the outside of the hollow fibers are isolated from each other. The container is made of metallic materials such as stainless steel, plastics, and composite materials such as fiber-reinforced plastics.

Detailed Description Text - DETX (17):

The separation **membrane** modules used in the present invention are not particularly limited in shape and can have a commonly employed shape. The hollow fibers are bundled in a parallel configuration, a crossing configuration, a woven configuration or a helical configuration. The hollow fiber bundle may have a core tube in substantially the center thereof. The outer peripheral surface of the hollow fiber bundle may be wrapped in a film. The shape of the hollow fiber bundle includes cylinders, flat plates, and prisms. The hollow fiber bundle can be placed in the container as such or as bent into a U-shape or as wound into a helical shape. The separation **membrane** modules used in the present invention may be of bore feed mode or shell feed mode.

Detailed Description Text - DETX (18):

The apparatus of the present invention for separating and recovering a

halide gas has at least two stage separation **membrane** modules. The first stage separation **membrane** module has its gas inlet connected to a mixed gas source containing a halide gas and its non-permeate gas outlet connected via a gas flow control valve to a gas recovery container. The second separation **membrane** module has its gas inlet connected to the permeate gas outlet of the first stage separation **membrane** module and its non-permeate outlet connected to the mixed gas source containing the halide gas.

Detailed Description Text - DETX (19):
In a preferred configuration of the halide gas separation and recovery apparatus of the present invention, the first stage separation **membrane** module has its non-permeate outlet connected to the gas recovery container via a gas concentration detector and the gas flow control valve. Still preferably, the gas concentration detector and the gas flow control valve are coupled by a controller so that the gas flow control valve is controlled based on a measured value of the gas concentration detector thereby to adjust the amount of the gas emitted from the non-permeate gas outlet of the first stage separation **membrane** module.

Detailed Description Text - DETX (20):
An illustrative embodiment of the apparatus according to the present invention is shown in FIG. 1. FIG. 1 illustrates the configuration of the apparatus according to the present invention. The present invention will be described in greater detail by referring to FIG. 1. In FIG. 1, numeral 11 indicates a mixed gas source containing a halide gas. The mixed gas source may be either a piece of such equipment as gas-insulated electric equipment, e.g., a gas-insulated switchgear, a gas circuit breaker or a gas-insulated transformer, or semiconductor fabrication equipment or a storage tank in which a halide gas-containing mixed gas emitted from such equipment is stored. The halide gas-containing mixed gas of the mixed gas source 11 is introduced through a conduit pipe into an apparatus 10 of the present invention. The part surrounded by the dotted line in FIG. 1 is the apparatus of the present invention. If needed, the mixed gas from the mixed gas source 11 may be led into the apparatus by means of a vacuum pump or a blower while not shown. The introduced mixed gas is pressurized in a compressor 12. Numerals 13 and 14 indicate a buffer tank and a pressure control valve, respectively, through which the mixed gas with a constant controlled pressure is fed to the gas inlet of a first stage separation **membrane** module 15. While the mixed gas flows in the first stage separation **membrane** module, the diluting gas in the mixed gas (e.g., N.sub.2 gas) is selectively allowed to pass through the gas separation **membrane**. On the other hand, having a lower rate of permeation than the diluting gas, the halide gas (e.g., SF.sub.6 gas) in the mixed gas is

concentrated and recovered through the non-permeate gas outlet, while partly allowed to pass through the gas separation **membrane**. The non-permeate gas outlet connects via a gas concentration detector 17 and a gas flow control valve 16 to a gas recovery container 18. The gas concentration detector 17 measures the concentration of the halide gas concentrated and discharged from the non-permeate gas outlet, and the gas flow control valve 16 controls the amount of the gas discharged from the non-permeate gas outlet. When the opening of the gas flow control valve 16 is narrowed to reduce the gas flow from the non-permeate gas outlet, the amount of the permeate gas (i.e., the diluting gas plus a relatively small amount of the halide gas) which permeates through the gas separation **membrane** increases. This results in an increase in concentration of the halide gas recovered from the non-permeate gas outlet. Conversely, when the gas flow control valve 16 is opened to increase the gas flow from the non-permeate gas outlet, the amount of the diluting gas which passes through the gas separation **membrane** decreases, and the amount of the diluting gas remaining in the non-permeate gas increases relatively. It follows that the concentration of the halide gas collected from the non-permeate gas outlet decreases.

Detailed Description Text - DETX (21):

Since the concentration of the halide gas collected from the non-permeate gas outlet of the first stage separation **membrane** module can thus be adjusted to a prescribed high concentration (high purity), it is easy to reuse the recovered gas as a halide gas. Because a highly concentrated halide gas is easy to liquefy, the recovered halide gas may be liquefied with ease by pressure cooling. It is advantageous that the gas recovery container 18 be adapted to liquefy the halide gas and to store the liquid. In this case, the size of the container 18 can be markedly reduced as compared with a design for storing gas. Where the gas is liquefied by, for example, pressure cooling and stored in the form of liquid, the liquefied product will have a further increased purity, which is advantageous in reusing the stored liquid.

Detailed Description Text - DETX (22):

On the other hand, the permeate gas of the first stage separation **membrane** module 15 comprises the diluting gas as concentrated and the halide gas as diluted. The permeate gas outlet of the first stage separation **membrane** module 15 is connected via a compressor 19 to the gas inlet of a second stage separation **membrane** module 22. The permeate gas of the first stage separation **membrane** module 15 is pressurized in the compressor 19 and fed to the gas inlet of the second stage separation **membrane** module 22. Numeral 20 indicates a control valve for controlling the flow in a conduit bypassing the compressor 19, and numeral 21 a buffer tank. By these two units, the pressure and the

flow of the gas fed to the second separation **membrane** module 22 can be adjusted. The non-permeate gas outlet of the second stage gas separation **membrane** module 22 is connected via a control valve 23 and a check valve 24 to the mixed gas source 11. The permeate gas outlet of the second stage separation **membrane** module 22 is connected to a recovery tank 25.

Detailed Description Text - DETX (23):

The halide gas of the mixed gas fed to the apparatus of the present invention can be separated and recovered with a prescribed high purity by controlling the gas flow by means of the gas flow control valve 16 which is connected to the non-permeate gas outlet of the first stage separation **membrane** module 15. From the permeate gas outlet of the first stage separation **membrane** module 15 is discharged a mixed gas comprising the diluting gas as concentrated and the halide gas that has permeated through the gas separation **membrane** together with the diluting gas. In particular, narrowing the opening of the gas flow control valve 16 to reduce the gas flow in order to separate and recover the halide gas with high purity as the non-permeate gas of the first stage separation **membrane** module 15 results in increase in amount of the halide gas that permeates the first stage separation **membrane** module 15 into the permeate side together with the diluting gas. In this case, emission of this permeate gas directly into the atmosphere would result in not only a failure to increase the recovery rate of the halide gas but also adverse influences on the global environment. In the apparatus of the present invention, the permeate gas from the first stage separation **membrane** module 15 is fed to the second stage separation **membrane** module 22, where it is again separated into the diluting gas and the halide gas. As a result, the permeate gas from the second stage separation **membrane** module 22 has a remarkably reduced halide gas content, and the diluting gas can be recovered with a higher concentration. The non-permeate gas from the second stage separation **membrane** module 22 contains the non-recovered halide gas. This non-permeate gas is recycled to the mixed gas source 11 and again fed to the first stage separation **membrane** module. Therefore, no recovery loss occurs, and an increased recovery rate of the halide gas can be obtained.

Detailed Description Text - DETX (24):

Steam is far more permeable through a gas separation **membrane** than halide gases and diluting gases. Even where the halide gas-containing mixed gas treated in the present invention contains steam, steam passes through the gas separation **membranes** with the diluting gas. As a result, the recovered halide gas is dry and suitable to be reused as an electrically insulating gas or a processing gas for semiconductor fabrication.

Detailed Description Text - DETX (25):

In a preferred configuration of the apparatus according to the present invention, the gas concentration detector 17 and the gas flow control valve 16 are coupled by a controller 26 as illustrated in FIG. 1. According to this configuration, the gas flow control valve 16 is controlled based on measured values of the gas concentration detector 17 so as to adjust the amount of the gas discharged from the non-permeate gas outlet. More specifically, the gas concentration detector 17 measures the concentration of the halide gas recovered as a non-permeate gas of the first stage separation **membrane** module 15. When the measured value is below a previously determined gas concentration, the controller 26 automatically narrows the opening of the gas flow control valve 16 to reduce the amount of the gas to be collected through the non-permeate gas outlet of the first stage separation **membrane** module 15 thereby to raise the recovered halide gas concentration to the predetermined level. When, on the other hand, the measured value is above the previously determined gas concentration, the controller 26 automatically opens the gas flow control valve 16 to increase the amount of the gas to be collected through the non-permeate gas outlet of the first stage separation **membrane** module 15 thereby to adjust the recovered halide gas concentration to the predetermined level. Such an automatic control system enables efficient separation and recovery of the halide gas with a predetermined high concentration.

Detailed Description Text - DETX (26):

The gas concentration detector 17 used in the present invention is not particularly limited as long as it is capable of detecting the concentration of a halide gas. Considering that the purpose of using the detector is to control the gas flow from the non-permeate gas outlet of the first stage separation **membrane** module, the detector is preferably such that is capable of rapidly providing measured values without requiring a long time for measurement. Detectors capable of converting measured values to electric signals are suitable. Such detectors include those utilizing difference in sound transmission velocity and those utilizing change in heat conductivity. The controller 26 can be of general type that carries out control through electrical signals. It may be of the type that sets a single value or of the type that sets two values, the upper and the lower limits. The gas flow control valve 16 can be of general type that opens and closes according to electrical signals.

Detailed Description Text - DETX (27):

Another illustrative embodiment of the apparatus according to the present invention is shown in FIG. 2. FIG. 2 illustrates the configuration of the apparatus of the present invention. In the embodiment shown in FIG. 2, the

permeate gas outlet of the second separation **membrane** module 22 is connected via a compressor 27 to the gas inlet of a third stage separation **membrane** module 30. Numeral 28 indicates a control valve for controlling the flow in a conduit bypassing the compressor 27, and numeral 29 a buffer tank. The permeate gas of the second stage separation **membrane** module 22 is sent to the third separation **membrane** module 30 while having its pressure and flow adjusted by the compressor 27, the control valve 28, the buffer tank 29, and a control valve 31 which is connected to the non-permeate gas outlet of the third stage separation **membrane** module 30. The non-permeate gas outlet of the third stage gas separation **membrane** module 30 is connected via the control valve 31 and a check valve 24 to the mixed gas source 11. According to the embodiment of FIG. 2, even where the permeate gas discharged from the second stage separation **membrane** module 22 still contains the halide gas in such a concentration that is not ignorable as a recovery loss or not acceptable from the considerations for the global environment, it is introduced into the third separation **membrane** module 30, where it is separated into the diluting gas and the halide gas. As a result, the diluting gas can be recovered in a higher concentration with a lower halide gas content as a permeate gas from the third stage separation **membrane** module 30. While the non-permeate gas of the third stage separation **membrane** module 30 still contains the halide gas, it is recycled to the mixed gas source 11 and again fed to the first stage gas separation **membrane** module. Therefore, the residual halide gas never becomes a recovery loss but contributes to an increase of the halide gas recovery rate.

Detailed Description Text - DETX (28):

It is possible for this apparatus of the present invention to have one or more additional separation **membrane** modules having the same structure as the third stage separation **membrane** module so that the amount of the halide gas in the permeate gas from the final stage separation **membrane** module may be reduced to a level ignorable as a recovery loss or acceptable for the global environment.

Detailed Description Text - DETX (29):

It is desirable in the present invention that the separation of gas be carried out while maintaining the separation **membrane** module at a constant temperature in a range of from 40 to 200.degree. C.

Detailed Description Text - DETX (30):

Gas separation performance of a gas separation **membrane** is influenced by temperature. Where the gas separation **membrane** is not kept at a constant temperature, for example, when the separation **membrane** module is exposed to the influence of external temperature, it would be necessary to control the gas

flow control valve so frequently with temperature changes, and it would be difficult to separate and recover a halide gas with a prescribed concentration (purity) at a prescribed recovery rate. In order to separate and recover a halide gas in a high concentration at a high recovery rate as aimed in the present invention, it is particularly important to maintain the separation **membrane** module at a constant temperature.

Detailed Description Text - DETX (31):

Because gas separation **membranes** generally show higher rates of gas permeation at higher temperatures, the separation treatment is preferably conducted at higher temperatures. Further, the separation **membrane** tends to adsorb a halide gas to reduce its separating performance after long-term use. Adsorption of a halide gas onto the separation **membrane** can be suppressed by maintaining the module at high temperatures. The temperature at which the separation **membrane** module is to be maintained is 40.degree. C. or higher, preferably 80.degree. C. or higher, still preferably 110.degree. C. or higher. On the other hand, where maintained at too high temperatures, the separation **membrane** module, inclusive of the separation **membrane** and other elements, must be so heat resistant, and high energy consumption will lead to bad economy. Hence, the temperature at which the module is to be maintained is 200.degree. C. or lower, preferably 180.degree. C. or lower, still preferably 150.degree. C. or lower.

Detailed Description Text - DETX (32):

In order to maintain the separation **membrane** module at a temperature within the above-recited range, the module is provided with a heating unit having a temperature control function. The heating unit comprises, for example, a temperature sensor, a heater, and a controller capable of controlling these tools.

Detailed Description Text - DETX (33):

In the apparatus of the present invention, the permeate gas discharged from the permeate gas outlet of the final stage separation **membrane** module mainly comprises the concentrated diluting gas. It is possible to reduce the halide gas content of this permeate gas to such a level that is acceptable as a recovery loss or for the global environment. Therefore, the permeate gas from the final stage is permitted to be emitted into the atmosphere as such. If desired, the final permeate gas may be subjected to treatment with an adsorbent before emission or once stored in a recovery tank and then subjected to an appropriate treatment.

Detailed Description Text - DETX (34):

If necessary, the apparatus of the present invention may be equipped with a dust filter, an oil separator, a mist separator, a scrubber, a thermometer, a pressure gauge, a unit for treatment with an adsorbent, a gas concentration analyzer, a reducing valve, a flow control valve, a heating unit, a cooling unit, a pressurizing unit, a vacuum pump, a tank, and the like. Where the halide gas-containing mixed gas contains small amounts of impurities (such as gases resulting from deterioration of a halide gas), the apparatus may be configured to remove such impurities before the mixed gas is fed to the first stage separation **membrane** module by means of a unit for treatment with an adsorbent or a scrubber. Where the mixed gas has an extremely low halide gas concentration, it may be preliminarily concentrated to raise its halide gas concentration to about 1% by volume, preferably about 3% by volume, before it is fed to the first stage separation **membrane** module. The halide gas-containing mixed gas may be once heated or cooled to a predetermined temperature and then supplied to the first stage separation **membrane** module.

Detailed Description Text - DETX (35):

The halide gas separation and recovery method according to the present invention is a method of separating and recovering a halide gas through multiple stages using at least two separation **membrane** modules, which is characterized by comprising the steps of feeding a mixed gas containing a halide gas to the gas inlet of a first stage separation **membrane** module, feeding, to the gas inlet of each of the second and the following stage separation **membrane** modules, a permeate gas of an immediately preceding stage separation **membrane** module, recycling the non-permeate gas of each of the second and the following stage separation **membrane** modules to the gas inlet of the first stage separation **membrane** module, and collecting the halide gas as a non-permeate gas of the first stage separation **membrane** module in a prescribed concentration while controlling the non-permeate gas flow from the first stage separation **membrane** module with a gas flow control valve connected to the non-permeate gas outlet of the first stage separation **membrane** module. This method makes it possible to arbitrarily adjust the concentration of the recovered halide gas. The halide gas that constitutes a recover loss is only the one present in the gas mainly comprising the diluting gas that is emitted from the permeate gas outlet of the final stage separation **membrane** module. It is possible to reduce a recovery loss to an extremely low level. That is, the method of the present invention enables separation and recovery of a halide gas in high concentrations (purity) at high recover rates.

Detailed Description Text - DETX (36):

Still preferably, the halide gas separation and recovery method according to the present invention is characterized in that a gas concentration detector and

the gas flow control valve are connected to the non-permeate gas outlet of the first stage separation **membrane** module. In this preferred method, the gas concentration detector measures the halide gas concentration of the non-permeate gas discharged from the non-permeate gas outlet, and the gas flow control valve controls the flow of the non-permeate gas based on the measured value of the gas concentration detector, whereby the non-permeate gas is recovered as a halide gas in a controlled concentration. According to this method, it is easy to adjust the concentration of the halide gas to be recovered to an arbitrarily decided concentration. In particular, it is easy to recover the halide gas in a high concentration (with a high purity) acceptable for reuse.

Detailed Description Text - DETX (37):

Taking reusability into consideration, the halide gas concentration in the non-permeate gas recovered from the first stage separation **membrane** module is suitably 90% by volume or higher, preferably 95% by volume or higher, still preferably 98% by volume or higher. In order to minimize adverse influences on the global environment, the recovery loss is suitably 10% or less, preferably 3% or less, still preferably 1% or less.

Detailed Description Text - DETX (42):

A gas-insulated electric apparatus having a capacity of 205 liters which was charged with a mixed gas of 10 vol % SF.sub.6 gas and 90 vol % N.sub.2 gas under a pressure of 0.25 MPa (absolute pressure, hereinafter the same) was used as a halide gas-containing mixed gas source 11. The mixed gas was pressurized in a compressor 12 and led via a pressure control valve 14 to the gas inlet of the first stage separation **membrane** module 15 under a pressure of 0.8 MPa. The first and second separation **membrane** modules 15 and 22 each had a large number of hollow fiber **membranes**. The hollow fiber **membranes** were aromatic polyimide **asymmetric** hollow fiber **membranes** prepared from 30 mmol, in terms of proportion (hereinafter the same), of 3,3',4,4'-biphenyltetracarboxylic acid dianhydride, 55 mmol of 2,2'-bis(3,4-dicarboxyphenyl)hexafluoropropane dianhydride, 15 mmol of pyromellitic dianhydride, 50 mmol of 3,7-diamino-2,8-dimethyldiphenylenesulfone, and 50 mmol of 2,2',5,5'-tetrachlorobenzidine. Each hollow fiber **membrane** had an outer diameter of 410 .mu.m and an inner diameter of 280 .mu.m. The hollow fiber **membranes** were gathered into a hollow fiber bundle, and both end portions of the bundle were firmly fixed with an epoxy resin. The bundle was set in a container having a gas inlet, a permeate gas outlet, and non-permeate gas outlet to make the separation **membrane** modules 15 and 22. These separation **membrane** modules 15 and 22 both had an effective **membrane** area of 2 m.sup.2.

Detailed Description Text - DETX (43):

Of the mixed gas components introduced into the separation **membrane** module 15, the gas that did not permeate through the hollow fiber separation **membrane** was made to flow from the non-permeate gas outlet of the separation **membrane** module 15 and led via a gas flow control valve 16 and a gas concentration detector 17 to a gas recovery container 18 and thus recovered. In this route, a controller 26 automatically controlled the gas flow control valve 16 to regulate the non-permeate gas flow so that the SF.sub.6 gas concentration measured in the gas concentration detector 17 might be 96 vol %. The permeate gas of the separation **membrane** module 15 flowing from the permeate gas outlet was adjusted to 0.4 MPa by a compressor 19 and led to the gas inlet of the second stage separation **membrane** module 22. The non-permeate gas of the second stage separation **membrane** module 22 flew from the non-permeate gas outlet and returned to the gas-insulated electric apparatus (mixed gas source 11) via a control valve 23 and a check valve 24. The permeate gas of the second stage separation **membrane** module 22 was led from the permeate gas outlet toward a recovery tank 25, where it was stored. Each of the separation **membrane** modules 15 and 22 was maintained at 100.degree. C. by the respective ribbon heaters wound therearound. Each of the gas recovery container 18 and the recovery tank 25 was used after purging with a vacuum pump.

Detailed Description Text - DETX (44):

After 40-minute operation under these conditions, the amount of the gas collected in the gas recovery container 18 from the non-permeate gas outlet of the first stage separation **membrane** module 15 was 44 liters, and its SF.sub.6 concentration was 96 vol %. The amount of the gas collected in the recovery tank 25 from the permeate gas outlet of the second stage separation **membrane** module was 400 liters, and its SF.sub.6 gas concentration was 0.2 vol %. That is, the recovery loss and the recovery rate of SF.sub.6 gas were 1.8% and 98.2%, respectively.

Detailed Description Text - DETX (47):

Similarly to Example 1, a gas-insulated electric apparatus having a capacity of 205 liters and charged with a mixed gas of 10 vol % SF.sub.6 gas and 90 vol % N.sub.2 gas under a pressure of 0.25 MPa was used as a halide gas-containing mixed gas source 11. The mixed gas was pressurized in the compressor 12 and led via the pressure control valve 14 to the gas inlet of the first stage separation **membrane** module 15 under a pressure of 0.8 MPa. Of the mixed gas components introduced into the separation **membrane** module 15, the gas that did not permeate through the hollow fiber separation **membrane** was made to flow from the non-permeate gas outlet of the separation **membrane** module 15 and led via the gas flow control valve 16 and the gas concentration detector 17 to the gas

recovery container 18 and thus recovered. In this route, the controller 26 automatically controlled the gas flow control valve 16 to regulate the non-permeate gas flow so that the SF.sub.6 gas concentration measured in the gas concentration detector 17 might be 99 vol %. The permeate gas of the separation **membrane** module 15 flowing from the permeate gas outlet was adjusted to 0.3 MPa by the compressor 19 and led to the gas inlet of the second stage separation **membrane** module 22. The non-permeate gas of the second stage separation **membrane** module 22 flew from the non-permeate gas outlet and returned to the gas-insulated electric apparatus (mixed gas source 11) via the control valve 23 and the check valve 24. The permeate gas of the second stage separation **membrane** module 22 was led from the permeate gas outlet toward the recovery tank 25, where it was stored. Each of the separation **membrane** modules 15 and 22 was maintained at 100.degree. C. by the respective ribbon heaters wound therearound. Each of the gas recovery container 18 and the recovery tank 25 was used after purging with a vacuum pump.

Detailed Description Text - DETX (48):

After 40-minute operation under these conditions, the amount of the gas collected in the gas recovery container 18 from the non-permeate gas outlet of the first stage separation **membrane** module 15 was 36 liters, and its SF.sub.6 concentration was 99 vol %. The amount of the gas collected in the recovery tank 25 from the permeate gas outlet of the second stage separation **membrane** module was 370 liters, and its SF.sub.6 concentration was 0.1 vol % or lower. That is, the recovery loss and the recovery rate of SF.sub.6 gas were 1.0% or less and 99.0% or higher, respectively.

Detailed Description Text - DETX (52):

Preparation of **Asymmetric** Hollow Fiber **Membrane**:

Detailed Description Text - DETX (53):

The polyimide solution was put into a spinning apparatus equipped with a hollow fiber spinning nozzle (outer diameter of annular opening: 800 .mu.m; width of annular opening: 100 .mu.m; outer diameter of core opening: 300 .mu.m). The polyimide solution was extruded through the spinning nozzle while feeding nitrogen gas through the core opening of the spinning nozzle. The spun hollow fiber was passed through a nitrogen atmosphere, immersed in a primary coagulant bath (80 wt % aqueous ethanol solution) at 0.degree. C., led into a secondary coagulant bath having guide rollers, where the fiber ran reciprocally as guided by the guide rollers to complete coagulation, and wound on a bobbin. The resulting aromatic polyimide hollow fiber **membrane** was thoroughly washed with ethanol. After exchanging the solvent for isooctane, the fiber was dried by heating at 100.degree. C. to evaporate isooctane and then heat-treated at

270.degree. C. for 30 minutes to give an aromatic polyimide **asymmetric** hollow fiber **membrane** having an outer diameter of about 240 .mu.m and an inner diameter of about 160 .mu.m.

Detailed Description Text - DETX (54):
Preparation of Partially Carbonized **Membrane**:

Detailed Description Text - DETX (55):
The aromatic polyimide **asymmetric** hollow fiber **membrane** was thermally is stabilized by preliminarily heating at 400.degree. C. for 30 minutes in air with no tension applied. The hollow fiber **membrane** was then passed through a nitrogen-filled quartz glass tube which was placed in an electric tube furnace the inside of which was kept at 600.degree. C. for a retention time of 4 minutes to obtain a partially carbonized **asymmetric** hollow fiber **membrane** having an outer diameter of about 200 .mu.m and an inner diameter of about 140 .mu.m.

Detailed Description Text - DETX (59):
Preparation of **Asymmetric** Hollow Fiber **Membrane**:

Detailed Description Text - DETX (60):
The polyimide solution was charged into a spinning apparatus equipped with a hollow fiber spinning nozzle (outer diameter of annular opening: 1000 .mu.m; width of annular opening: 200 .mu.m; outer diameter of core opening: 400 .mu.m). The polyimide solution was processed in the same manner as in Reference Example 1, except for using a 70 wt % aqueous ethanol solution as a coagulant bath, to obtain an aromatic polyimide **asymmetric** hollow fiber **membrane** having an outer diameter of about 360 .mu.m and an inner diameter of about 270 .mu.m.

Detailed Description Text - DETX (61):
Preparation of Partially Carbonized **Membrane**:

Detailed Description Text - DETX (62):
The aromatic polyimide **asymmetric** hollow fiber **membrane** was thermally stabilized by preliminarily heating at 400.degree. C. for 30 minutes in air with no tension applied. The hollow fiber **membrane** was then passed through a nitrogen-filled quartz glass tube which was placed in an electric tube furnace the inside of which was kept at 600.degree. C. for a retention time of 4 minutes to obtain a partially carbonized **asymmetric** hollow fiber **membrane** having an outer diameter of about 320 .mu.m and an inner diameter of about 240 .mu.m.

Detailed Description Text - DETX (65):

Each of the first stage separation **membrane** module 15 and the second stage separation **membrane** module 22 was prepared by bundling a large number of hollow fiber **membranes** having an outer diameter of about 200 .mu.m and an inner diameter of about 140 .mu.m which were prepared in Reference Example 1, securing both end portions of the bundle with an epoxy resin, and setting the bundle in a container having a gas inlet, a permeate gas outlet, and a non-permeate gas outlet. The separation **membrane** modules 15 and 22 had an effective **membrane** area of 2.2 m.sup.2 and 0.65 m.sup.2, respectively.

Detailed Description Text - DETX (66):

A 205-liter volume tank charged with a mixed gas of 10 vol % CF.sub.4 gas and 90 vol % N.sub.2 gas under a gas pressure of 0.25 MPa was used as a halide gas-containing mixed gas source 11. The mixed gas was pressurized in the compressor 12 and led via the pressure control valve 14 to the gas inlet of the first stage separation **membrane** module 15 under a pressure of 0.9 MPa. Of the mixed gas components introduced into the separation **membrane** module 15, the gas that did not permeate through the hollow fiber separation **membrane** was made to flow from the non-permeate gas outlet of the separation **membrane** module 15 and led via the gas flow control valve 16 and the gas concentration detector 17 to the gas recovery container 18 and thus recovered. In this route, the controller 26 automatically controlled the gas flow control valve 16 to regulate the non-permeate gas flow so that the CF.sub.4 gas concentration measured in the gas concentration detector 17 might be 99 vol %. The permeate gas of the separation **membrane** module 15 flowing from the permeate gas outlet was adjusted to 0.5 MPa by the compressor 19 and led to the gas inlet of the second stage separation **membrane** module 22. The non-permeate gas of the second stage separation **membrane** module 22 flew from the non-permeate gas outlet and returned to the tank (mixed gas source 11) via the control valve 23 and the check valve 24. The permeate gas of the second stage separation **membrane** module 22 was led from the permeate gas outlet toward the recovery tank 25, where it was stored. Each of the separation **membrane** modules 15 and 22 was maintained at 100.degree. C. by the respective ribbon heaters wound therearound. Each of the gas recovery container 18 and the recovery tank 25 was used after purging with a vacuum pump.

Detailed Description Text - DETX (67):

After 40-minute operation under these conditions, the amount of the gas collected in the gas recovery container 18 from the non-permeate gas outlet of the first stage separation **membrane** module 15 was 39 liters, and its CF.sub.4 gas concentration was 99 vol %. The amount of the gas collected in the recovery

tank 25 from the permeate gas outlet of the second stage separation **membrane** module was 391 liters, and its CF.sub.4 gas concentration was 0.1 vol % or lower. That is, the recovery loss and the recovery rate of CF.sub.4 gas were 1.0% or less and 99% or higher, respectively.

Detailed Description Text - DETX (70):

A 205-liter volume tank having charged therein a mixed gas of 10 vol % NF.sub.3 gas and 90 vol % N.sub.2 gas under a gas pressure of 0.25 MPa was used as a halide gas-containing mixed gas source 11. The mixed gas was pressurized in the compressor 12 and led via the pressure control valve 14 to the gas inlet of the first stage separation **membrane** module 15 under a pressure of 0.5 MPa. Of the mixed gas components introduced into the separation **membrane** module 15, the gas that did not permeate through the hollow fiber separation **membrane** was made to flow from the non-permeate gas outlet of the separation **membrane** module 15 and led via the gas flow control valve 16 and the gas concentration detector 17 to the gas recovery container 18 and thus recovered. In this route, the controller 26 automatically controlled the gas flow control valve 16 to regulate the non-permeate gas flow so that the NF.sub.3 gas concentration measured in the gas concentration detector 17 might be 99 vol %. The permeate gas of the separation **membrane** module 15 flowing from the permeate gas outlet was adjusted to 0.6 MPa by the compressor 19 and led to the gas inlet of the second stage separation **membrane** module 22. The non-permeate gas of the second stage separation **membrane** module 22 flew from the non-permeate gas outlet and returned to the tank (mixed gas source 11) via the control valve 23 and the check valve 24. The permeate gas of the second stage separation **membrane** module 22 was led from the permeate gas outlet toward the recovery tank 25, where it was stored. Each of the separation **membrane** modules 15 and 22 was maintained at 50.degree. C. by the respective ribbon heaters wound therearound. Each of the gas recovery container 18 and the recovery tank 25 was used after purging with a vacuum pump.

Detailed Description Text - DETX (71):

After 40-minute operation under these conditions, the amount of the gas collected in the gas recovery container 18 from the non-permeate gas outlet of the first stage separation **membrane** module 15 was 34 liters, and its NF.sub.3 gas concentration was 99 vol %. The amount of the gas collected in the recovery tank 25 from the permeate gas outlet of the second stage separation **membrane** module was 289 liters, and its NF.sub.3 gas concentration was 0.2 vol. % or lower. That is, the recovery loss and the recovery rate of NF.sub.3 gas were 1.7% or less and 98.3% or higher, respectively.

Detailed Description Text - DETX (74):

Each of the first stage separation **membrane** module 15 and the second stage separation **membrane** module 22 was prepared in the same manner as in Example 3, except for using a large number of hollow fiber **membranes** having an outer diameter of about 320 .mu.m and an inner diameter of about 240 .mu.m which were prepared in Reference Example 2. The separation **membrane** modules 15 and 22 had an effective **membrane** area of 2.2 m.sup.2 and 0.65 m.sup.2, respectively.

Detailed Description Text - DETX (75):

A 205-liter volume tank having charged therein a mixed gas of 10 vol % SF.sub.6 gas and 90 vol % N.sub.2 gas under a gas pressure of 0.25 MPa was used as a halide gas-containing mixed gas source 11. The mixed gas was pressurized in the compressor 12 and led via the pressure control valve 14 to the gas inlet of the first stage separation **membrane** module 15 under a pressure of 0.8 MPa. Of the mixed gas components introduced into the separation **membrane** module 15, the gas that did not permeate through the hollow fiber separation **membrane** was made to flow from the non-permeate gas outlet of the separation **membrane** module 15 and led via the gas flow control valve 16 and the gas concentration detector 17 to the gas recovery container 18 and thus recovered. In this route, the controller 26 automatically controlled the gas flow control valve 16 to regulate the non-permeate gas flow so that the SF.sub.6 gas concentration measured in the gas concentration detector 17 might be 99 vol %. The permeate gas of the separation **membrane** module 15 flowing from the permeate gas outlet was adjusted to 0.4 MPa by the compressor 19 and led to the gas inlet of the second stage separation **membrane** module 22. The non-permeate gas of the second stage separation **membrane** module 22 flew from the non-permeate gas outlet and returned to the tank (mixed gas source 11) via the control valve 23 and the check valve 24. The permeate gas of the second stage separation **membrane** module 22 was led from the permeate gas outlet toward the recovery tank 25, where it was stored. Each of the separation **membrane** modules 15 and 22 was maintained at 100.degree. C. by the respective ribbon heaters wound therearound. Each of the gas recovery container 18 and the recovery tank 25 was used after purging with a vacuum pump.

Detailed Description Text - DETX (76):

After 40-minute operation under these conditions, the amount of the gas collected in the gas recovery container 18 from the non-permeate gas outlet of the first stage separation **membrane** module 15 was 22 liters, and its SF.sub.6 gas concentration was 99 vol %. The amount of the gas collected in the recovery tank 25 from the permeate gas outlet of the second stage separation **membrane** module was 215 liters, and its SF.sub.6 gas concentration was 0.1 vol % or lower. That is, the recovery loss and the recovery rate of SF.sub.6 gas were 1.0% or less and 99.0% or higher, respectively.

Detailed Description Text - DETX (79):

A 205-liter volume tank having charged therein a mixed gas of 10 vol % C.sub.2 F.sub.6 gas and 90 vol % N.sub.2 gas under a gas pressure of 0.25 MPa was used as a halide gas-containing mixed gas source 11. The mixed gas was pressurized in the compressor 12 and led via the pressure control valve 14 to the gas inlet of the first stage separation **membrane** module 15 under a pressure of 0.8 MPa. Of the mixed gas components introduced into the separation **membrane** module 15, the gas that did not permeate through the hollow fiber separation **membrane** was made to flow from the non-permeate gas outlet of the separation **membrane** module 15 and led via the gas flow control valve 16 and the gas concentration detector 17 to the gas recovery container 18 and thus recovered. In this route, the controller 26 automatically controlled the gas flow control valve 16 to regulate the non-permeate gas flow so that the C.sub.2 F.sub.6 gas concentration measured in the gas concentration detector 17 might be 99 vol %. The permeate gas of the separation **membrane** module 15 flowing from the permeate gas outlet was adjusted to 0.4 MPa by the compressor 19 and led to the gas inlet of the second stage separation **membrane** module 22. The non-permeate gas of the second stage separation **membrane** module 22 flew from the non-permeate gas outlet and returned to the tank (mixed gas source 11) via the control valve 23 and the check valve 24. The permeate gas of the second stage separation **membrane** module 22 was led from the permeate gas outlet toward the recovery tank 25, where it was stored. Each of the separation **membrane** modules 15 and 22 was maintained at 120.degree. C. by the respective ribbon heaters wound therearound. Each of the gas recovery container 18 and the recovery tank 25 was used after purging with a vacuum pump.

Detailed Description Text - DETX (80):

After 35-minute operation under these conditions, the amount of the gas collected in the gas recovery container 18 from the non-permeate gas outlet of the first stage separation **membrane** module 15 was 44 liters, and its C.sub.2 F.sub.6 gas concentration was 99 vol %. The amount of the gas collected in the recovery tank 25 from the permeate gas outlet of the second stage separation **membrane** module was 402 liters, and its C.sub.2 F.sub.6 gas concentration was 0.1 vol % or lower. That is, the recovery loss and the recovery rate of C.sub.2 F.sub.6 gas were 1.0% or less and 99.0% or higher, respectively.

Detailed Description Text - DETX (81):

Comparative Examples 1 to 3 are shown below. In Comparative Examples 1 to 3, a halide gas was separated and recovered with a purity of 99 vol % as a non-permeate gas in the same apparatus as used in Examples 3 to 5, except for using only the first but not the second stage separation **membrane** module. The

recovery losses were much greater than in Examples, reaching 10% or even higher.

Detailed Description Text - DETX (83):

In the same apparatus as used in Example 3, the permeate gas outlet of the first stage separation **membrane** module 15 was connected directly to the recovery tank 25. The same mixed gas as in Example 3 was treated in this apparatus by using only the first but not the second stage separation **membrane** module under the same separation conditions as in Example 3. The non-permeate gas was collected in the gas recovery container 18, and the permeate gas in the recovery tank 25.

Detailed Description Text - DETX (86):

In the same apparatus as used in Example 4, the permeate gas outlet of the first stage separation **membrane** module 15 was connected directly to the recovery tank 25. The same mixed gas as in Example 4 was treated in this apparatus using only the first but not the second stage separation **membrane** module under the same separation conditions as in Example 4. The non-permeate gas was collected in the gas recovery container 18, and the permeate gas in the recovery tank 25.

Detailed Description Text - DETX (89):

In the same apparatus as used in Example 5, the permeate gas outlet of the first stage separation **membrane** module 15 was connected directly to the recovery tank 25. The same mixed gas as used in Example 5 was treated in this apparatus using only the first but not the second stage separation **membrane** module under the same separation conditions as in Example 5. The non-permeate gas was collected in the gas recovery container 18, and the permeate gas in the recovery tank 25.

Detailed Description Paragraph Equation - DEEQ
(1):

Recovery loss (%)=(amount of halide gas in permeate gas of the final stage separation **membrane** module)/(amount of halide gas in non-permeate gas of the first stage separation **membrane** module+amount of halide gas in permeate gas of the final stage separation **membrane** module).times.100

Claims Text - CLTX (1):

1. A method of separating and recovering a halide gas from a mixed gas containing the halide gas through multiple stages using at least two separation **membrane** modules, which is characterized by comprising the steps of: feeding a mixed gas containing a halide gas to a gas inlet of a first stage separation

membrane module; feeding, to a gas inlet of each of a second and any following stage separation **membrane** modules, a permeate gas of an immediately preceding stage separation **membrane** module; recycling a non-permeate gas of each of the second and any following stage separation **membrane** modules to the gas inlet of the first stage separation **membrane** module; and collecting a non-permeate gas of the first stage separation **membrane** module with a prescribed halide gas concentration while controlling the non-permeate gas flow from the first stage separation **membrane** module with a gas flow control valve connected to the non-permeate gas outlet of the first stage separation **membrane** module.

Claims Text - CLTX (2):
2. The method of separating and recovering a halide gas according to claim 1, wherein a gas concentration detector and the gas flow control valve are connected to the non-permeate gas outlet of the first stage separation **membrane** module in such a configuration that the gas concentration detector measures the halide gas concentration of the non-permeate gas discharged from the non-permeate gas outlet of the first stage separation **membrane** module and that the gas flow control valve controls the non-permeate gas flow based on the measured value of the gas concentration detector thereby to collect the halide gas in a prescribed concentration.

Claims Text - CLTX (3):
3. The method of separating and recovering a halide gas according to claim 1, wherein each of the separation **membrane** modules is configured to be maintained at a constant temperature in a range of from 40 to 200.degree. C. by a heating unit.

Claims Text - CLTX (4):
4. The method of separating and recovering a halide gas according to claim 1, wherein each of the separation **membrane** modules comprises an aromatic polyimide hollow fiber separation **membrane**.

Claims Text - CLTX (5):
5. The method of separating and recovering a halide gas according to claim 1, wherein each of the separation **membrane** modules comprises a carbonized separation **membrane** obtained by carbonizing a polymer.

Claims Text - CLTX (6):
6. The method of separating and recovering a halide gas according to claim 5, wherein the carbonized separation **membrane** is a partially carbonized hollow fiber separation **membrane** obtained by partially carbonizing an aromatic polyimide **asymmetric** hollow fiber **membrane**.

Claims Text - CLTX (7):

7. An apparatus for separating and recovering a halide gas comprising at least first and second stage separation **membrane** modules which is characterized in that the first stage separation **membrane** module has a gas inlet thereof connected to a source of a mixed gas containing a halide gas and a non-permeate gas outlet thereof connected via a gas flow control valve to a gas recovery container, and the second stage separation **membrane** module has a gas inlet thereof connected to the permeate gas outlet of the first separation **membrane** module and a non-permeate gas outlet thereof connected to the source of the halide gas-containing mixed gas.

Claims Text - CLTX (8):

8. The apparatus for separating and recovering a halide gas according to claim 7, wherein the non-permeate gas outlet of the first stage separation **membrane** module is connected via the gas flow control valve and a gas concentration detector to the gas recovery container.

Claims Text - CLTX (10):

10. The apparatus for separating and recovering a halide gas according to claim 7, wherein each of the separation **membrane** modules is configured to be maintained at a constant temperature in a range of from 40 to 200.degree. C. by a heating unit.

Claims Text - CLTX (11):

11. The apparatus for separating and recovering a halide gas according to claim 7, wherein the at least first and second stage separation **membrane** modules comprise at least first, second and third stage separation **membrane** modules, wherein at least said third stage separation **membrane** module has a gas inlet thereof connected to a permeate gas outlet of an immediately preceding separation **membrane** module and a non-permeate gas outlet thereof connected to the source of the halide gas-containing mixed gas.

Claims Text - CLTX (12):

12. The apparatus for separating and recovering a halide gas according to claim 7, wherein each of the separation **membrane** modules comprises an aromatic polyimide hollow fiber separation **membrane**.

Claims Text - CLTX (13):

13. The apparatus for separating and recovering a halide gas according to claim 7, wherein each of the separation **membrane** modules comprises a carbonized separation **membrane** obtained by carbonizing a polymer.

Claims Text - CLTX (14):

14. The apparatus for separating and recovering a halide gas according to claim 13, wherein the carbonized separation **membrane** is a partially carbonized hollow fiber separation **membrane** obtained by partially carbonizing an aromatic polyimide **asymmetric** hollow fiber **membrane**.